

SOURCES AND DISTRIBUTION OF NO_x IN THE UPPER TROPOSPHERE AT NORTHERN MIDLATITUDES

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ABSTRACT

A simple quasi 2-D model is used to study the zonal distribution of NO_x. The model includes vertical transport in form of eddy diffusion and deep convection, zonal transport by a vertically uniform wind, and a simplified chemistry of NO, NO₂ and HNO₃. The NO_x sources considered are surface emissions (mostly from the combustion of fossil fuel), lightning, aircraft emissions, and downward transport from the stratosphere. The model is applied to the latitude band of 40°N to 50°N during the month of June; the contributions to the zonal NO_x distribution from the individual sources and transport processes are investigated. The model predicted NO_x concentration in the upper troposphere is dominated by air lofted from the polluted planetary boundary layer over the large industrial areas of Eastern North America and Europe. Aircraft emissions are also important and contribute on average 30 %. Stratospheric input is minor about 10 %, less even than that by lightning. The model provides a clear indication of intercontinental transport of NO_x and HNO₃ in the upper troposphere. Comparison of the modelled NO profiles over the Western Atlantic with those measured during STRATOZ III in 1984 shows good agreement at all altitudes.

1. INTRODUCTION

A major fraction of the available measurements of NO in the upper troposphere were made during the Stratospheric Ozone aircraft campaign STRATOZ III in June 1984 (Drummond et al., 1988). The flight track of that campaign was placed mainly along the coastlines of North America, South America, western North Africa and Europe between 70°N and 60°S latitude and from 0 to 12 km altitude. The most conspicuous feature in the individual vertical profiles (see for example figure 1) and in the two dimensional representation of all measurements (figure 2) is the large hump of high NO-Concentrations in the upper troposphere at 10-11 km altitude extending all over the northern hemisphere.

To explain this result, all possible NO-sources must be considered, namely stratospheric input, emissions by high flying aircraft, lightning and fast vertical transport from the planetary boundary layer. Unfortunately, the observed patterns in the NO-distribution were by themselves not sufficient to provide a quantitative estimate of the contribution of the various sources. Also the hope of using correlations with the concentrations of other trace gases measured during STRATOZ III, like O₃ and CO, proved vain so far. So we attempted to use a simple quasi

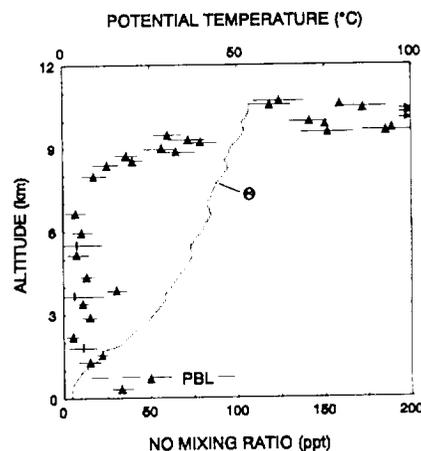


Fig. 1 Vertical profile of NO measured during STRATOZ III near Halifax

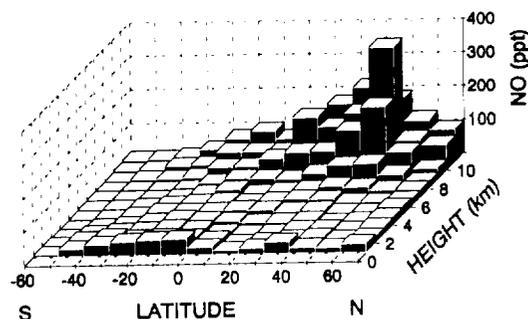


Fig. 2 Meridional representation of the average NO mixing ratios measured during STRATOZ III

2-D model to quantify the impact of the sources mentioned. The present paper summarizes the salient findings of our earlier work (Ehhalt et al., 1992).

2. MODEL DESCRIPTION

The model includes three types of transport : vertical transport by deep convection and eddy diffusion, and horizontal transport by vertically uniform wind. The fast vertical transport induced by deep convection is treated like a random process and superimposed onto a continuous transport by eddy diffusion. Selected by a random number, a fixed fraction of air at a specific altitude level is replaced by air from the boundary layer followed by a downward shift from each of the lower levels to the next one to maintain mass balance. This random process is adjusted such that the product of transport frequency and exchanged air fraction matches the exchange rate profiles (fig. 3) taken from the convection statistics of the GCM developed at the Goddard Institute of Space Studies (M. Prather, private communication 1990).

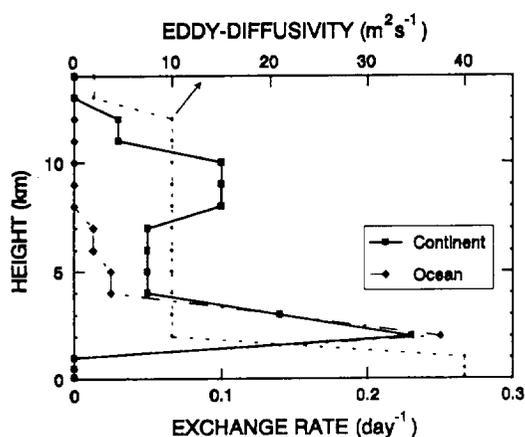
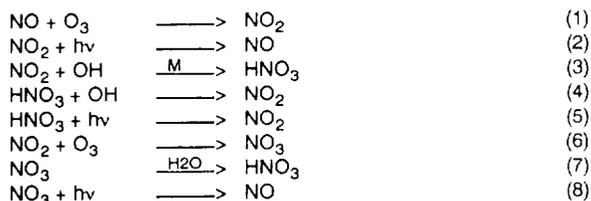


Fig. 3 Vertical profiles of the exchange rates of the fast vertical transport and of eddy diffusivity

Horizontal transport is introduced by moving the whole air column (0-14 km altitude) with a horizontal displacement given by a 8 ms^{-1} westerly wind (Houghton, 1985). The tropopause is located at 12 km altitude. Temperature and density profiles used are those of the U.S. Standard Atmosphere (1976). All parameters apply to the summer month at 40° - 50° N latitude.

The model allows the nitrogen compounds $\text{NO}_x = \text{NO} + \text{NO}_2$ and HNO_3 to be transported. Those species are interlinked by reactions (1) to (8).



The vertical concentration profiles of OH (Volz et al., 1981) and O_3 (Marengo and Said, 1989) and of the photolysis rates (Roeth, 1986) are held fixed at their diurnally averaged values. The rate constants used are those in Jet Propulsion Laboratory (JPL 1990).

The nitrogen compounds are removed from the atmosphere by dry and wet deposition. HNO_3 is deposited on all surfaces with a deposition velocity of 2 cm s^{-1} (Huebert and

Robert, 1985), NO_2 only on land surfaces with 0.5 cm s^{-1} (Boettger et al., 1978). Wet deposition of HNO_3 occurs in two modes. One is associated with the fast vertical transport events which are thought to proceed through convective clouds removing half of the HNO_3 lofted by the event. Additionally, HNO_3 is removed with a time constant of 10 days at altitudes of 1-12 km and with a time constant of 2 days below 1 km.

The emission rates of NO_x listed in Table 1 were calculated from Ehhalt and Drummond (1988) and Ehhalt and Drummond (1982). The emissions by lightning were coupled to the fast vertical transport by convective clouds allowing NO_x -injection for transport events which exceeded 8 km over the continents and 4 km over the oceans. The total emission rate by lightning was adjusted to match the value given in table 1.

Table 1 : NO_x Emissions in 40° - 50° N, June 1984

Source	Emission Rate 10^6 t N/yr
Surface source	
total	8.63
fossil fuel burning	7.45
Lightning	0.29
Aircraft, civil	0.081
Stratosphere	0.037

The aircraft emissions were derived from an average emission index of $10 \text{ g NO}_2/\text{kg fuel}$ and a fuel consumption in 1984 of $113 \cdot 10^6 \text{ t fuel/yr}$ (Nuesser and Schmitt, 1990). The vertical and longitudinal distribution was adopted from the Climatic Impact Assessment Program (CIAP, 1975).

3. RESULTS

The model was allowed to cycle three times around the globe to assume steady state. The zonal two dimensional distribution of NO and two sequences of vertical profiles of NO and NO_y are shown in figures 4 and 5. The contributions of the various sources in figure 5 were derived by model runs in which the individual source was switched off. Both figures demonstrate the development of a maximum in NO concentration at 10 km altitude as the air column passes over strong continental source areas. This maximum is a result of high surface concentrations carried upward by the fast vertical transport process. When the air column moves from a high surface source area out over the ocean (figure 5b), NO and NO_y are very quickly removed from the lower troposphere, but remain in the upper troposphere due to their long lifetime of about 10 days at those altitudes.

These results were tested against experimental results from Drummond et al. (1988). Figure 6 shows the comparison of the model predictions for the vertical NO-profiles at 50° W, 40° - 50° N and an average of the NO measurements between 50° N and 40° N about the south bound leg of STRAT0Z III. The two profiles agree surprisingly well indicating that all four sources are needed to produce the high NO-concentrations observed in the upper troposphere.

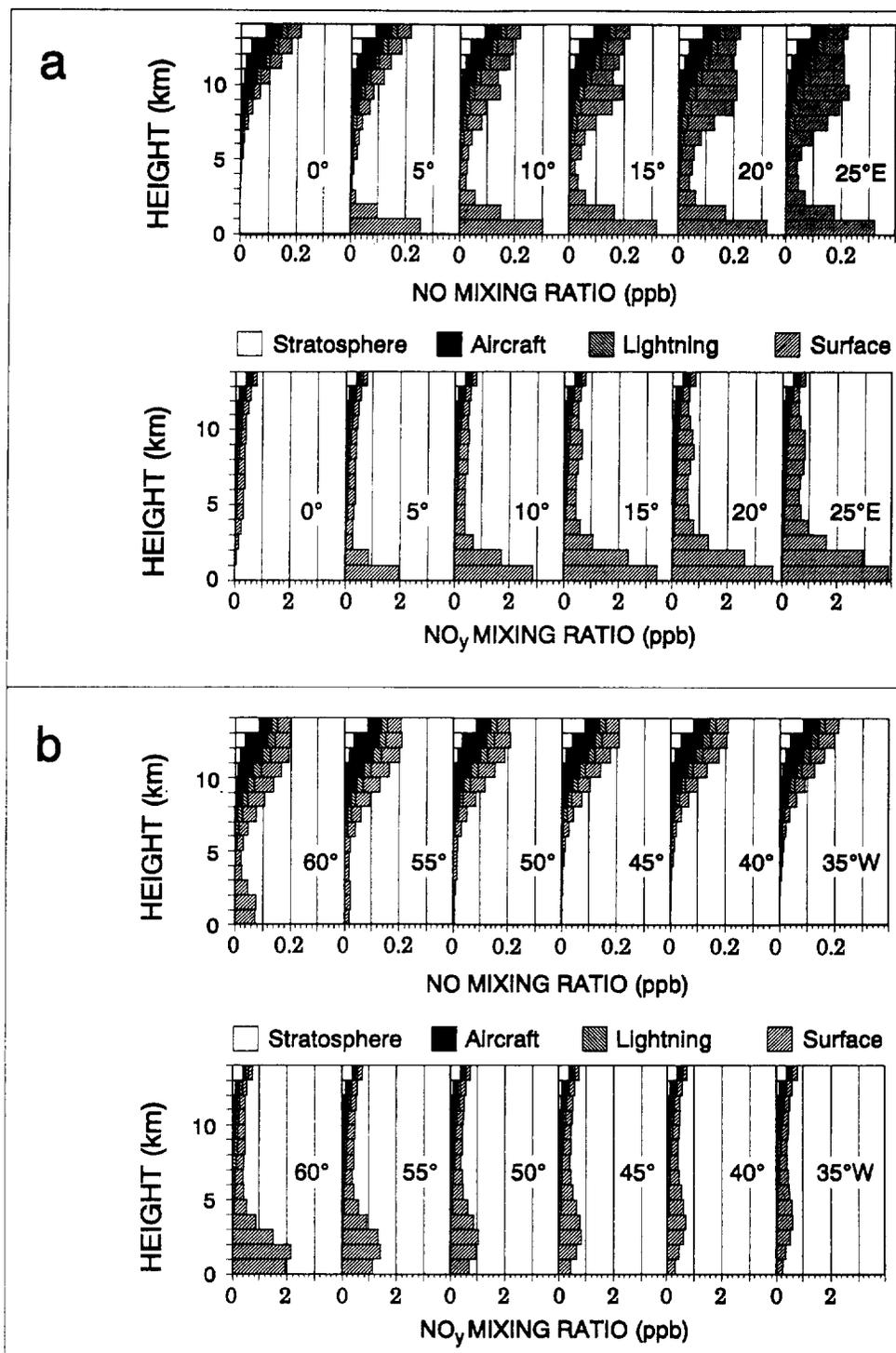


Fig 5 Calculated vertical mixing ratio profiles of NO and NO_y.
 a : over continental western Europe
 b : over the western North Atlantic

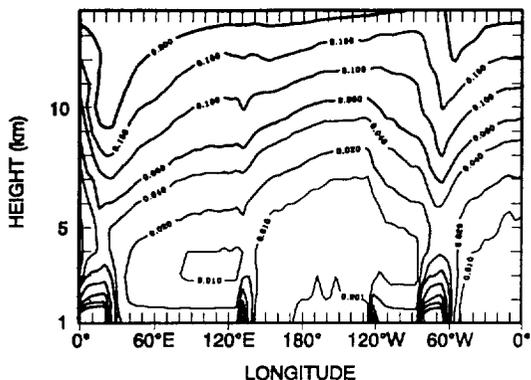


Fig. 4 Contour plot of the zonal two dimensional distribution of NO at 40°-50°N. Contours in ppb at 0.001, 0.01, 0.02, 0.04, 0.06, 0.1, 0.15 and 0.2

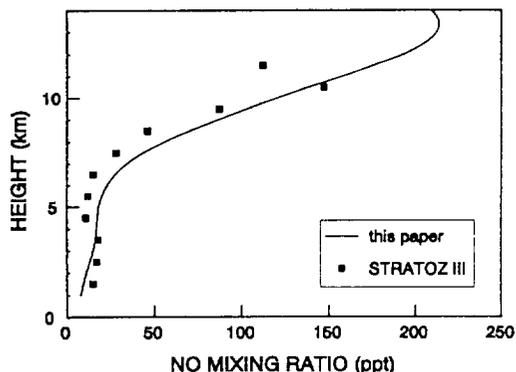


Fig. 6 Vertical profiles of the NO mixing ratio near 50°W, 40°-50°N, solid curve : model calculation, solid squares : averaged profiles measured during STRAT0Z III

4. CONCLUSIONS

Despite its simplicity the model used is capable of obtaining results similar to the salient features of the observed vertical NO-profiles at 40°-50°N in June 1984. In particular, it reproduces the high values and relative maxima in the vertical NO-profiles in the upper troposphere over coastal areas. The reason for this may be that for these latitudes and this time of year processes are indeed simple. The model facilitates a first examination of the contributions from various sources to the NO_x-concentration in the upper troposphere. Aircraft emissions which contribute on average about 30% and NO_x lofted from the planetary boundary layer are the major sources of NO_x in the upper troposphere at northern mid-latitudes during summer. The quantitative conclusions are of course uncertain. They depend on the assumed source strength which have errors of the order of a factor of 2. Work which has been done by using an improved 2-D model showed that the qualitative and quantitative conclusions given here did not depend on the assumption of vertically uniform wind speed or on the lack of horizontal eddy diffusion.

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